

Quantum Theory of Orbital Magnetization and its Generalization to Interacting Systems

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Based on standard perturbation theory, we present a full quantum derivation of the formula for the orbital magnetization in periodic systems. The derivation is generally valid for insulators with or without a Chern number, for metals at zero or finite temperatures, and at weak as well as strong magnetic fields. The formula is shown to be valid in the presence of electron-electron interaction, provided the one-electron energies and wave functions are calculated self-consistently within the framework of the exact current and spin density functional theory.

Magnetism is one of the most important properties of materials. Both spin and orbital motion of electrons can contribute to the total magnetization. While the spin magnetization can already be calculated from first principles with high accuracy by state-of-art methods such as the spin density functional theory (SDFT), the study of orbital magnetization is still in a comparatively primitive stage.

A first difficulty arises from the fact that there is still no theoretically well established formula for calculating the orbital magnetization of a crystalline solid. The non-locality of the orbital magnetization operator $\hat{\mathbf{M}} = -e\hat{\mathbf{r}} \times \hat{\mathbf{v}}$ is the major obstacle to obtaining a closed formula for an extended periodic system. Recently, Xiao et al. [1] and, independently, Thonhauser *et al.* [4, 5] obtained an orbital magnetization formula which avoids the non-locality problem and looks very promising for applications. However, up to date, there exists no general quantum mechanical derivation of this formula. The derivation presented in Ref. [1] relies on the semi-classical wave-packet dynamics of Bloch electrons [1, 2, 3], and its validity in the quantum context is not completely clear. On the other hand, the derivation presented in Ref. [4] is quantum mechanical, but relies on the existence of localized Wannier functions, and cannot be easily generalized to metals or insulators with non-zero Chern number. In addition, both derivations are limited to non-interacting systems. The shortcomings of these approaches call for a full quantum mechanical and many-body theory of the orbital magnetization.

A second difficulty is that a first principle calculation of the orbital magnetization (taking into account many-body effects) should be based on the spin current density functional theory (SCDFT) [7], rather than the conventional SDFT. Unfortunately, SCDFT has been hindered so far by the lack of reliable expressions for the magnetization-dependent effective potentials. This may partly explain why the orbital moments of ferromagnetic transition metals such as Fe, Co, and Ni calculated in SCDFT were found to be significantly smaller than the experimentally determined values [11]. How problematic these calculations are is well explained in the review article by Richter [12]. The situation, however, has been

rapidly changing in recent years. The advent of optimized effective potentials [13, 14] which treat exchange exactly and may systematically be improved for correlations opens new avenues to the study of magnetic materials. Applications to atoms and molecules have already appeared in the literature, and applications to periodic systems are the obvious next step.

Against this background, the present Letter serves a dual purpose. First, we present a general derivation of orbital magnetization in periodic systems based on the standard perturbation theory of quantum mechanics. The derivation clarifies the origin of the novel aspects of the semi-classical derivation, such as the Berry phase correction to the density of states. It is generally valid for metals and insulators with or without a Chern number, at zero or finite temperatures, in weak or strong magnetic field, and, of course, in the presence of spin-orbit coupling. Second, and most important, we combine this derivation with the exact current and spin density functional theory [6, 7], proving the validity of the magnetization formula for interacting systems. We believe that the magnetization formula, in combination with the recent advances the construction of optimized effective potential for SCDFT, will turn out to be a powerful practical tool for the study of systems that have long defied traditional ab-initio methods. [12]

We start from the standard thermodynamic definition of the orbital magnetization density:

$$\mathbf{M} = -\frac{1}{V} \left(\frac{\partial \Omega}{\partial \mathbf{B}} \right)_{T, \mu}, \quad (1)$$

where $\Omega = E - TS - \mu N$ is the grand thermodynamic potential, V is the total volume of the system, and \mathbf{B} is a magnetic field that only couples to the orbital motion of electrons (but does not contribute to the Zeeman energy) [18]. For convenience of derivation, we will first calculate the auxiliary quantity

$$\tilde{\mathbf{M}} \equiv -\frac{1}{V} \left(\frac{\partial K}{\partial \mathbf{B}} \right)_{T, \mu}, \quad (2)$$

where $K = E - \mu N$. We have $\mathbf{M} = \tilde{\mathbf{M}} + T(\partial S / \partial \mathbf{B})_{T, \mu}$ and, making use of the Maxwell relation $(\partial S / \partial \mathbf{B})_{T, \mu} =$

$(\partial \mathbf{M} / \partial T)_{\mu, \mathbf{B}}$, we have the simple relation back to the orbital magnetization:

$$\frac{\partial(\beta \mathbf{M})}{\partial \beta} = \tilde{\mathbf{M}}, \quad (3)$$

where $\beta \equiv 1/kT$.

In principle, one can evaluate $\tilde{\mathbf{M}}$ by employing the standard perturbation theory of quantum mechanics to calculate the energy correction due to a uniform magnetic field. However, such an approach will again hit the difficulty of the non-locality of the orbital magnetization operator. To go around the difficulty, we apply an external magnetic field that has an infinitely slow spatial variation [8, 9]:

$$\mathbf{B}(\mathbf{r}) = B \cos(qy) \hat{z} \quad (4)$$

The slow spatial variation of the field is controlled by the wave vector q , which will tend to zero at the end of the calculation. The correction to the energy density in this situation can be written, up to linear order in B , as

$$\delta K(\mathbf{r}) = -\tilde{\mathbf{M}} \cdot \mathbf{B}(\mathbf{r}). \quad (5)$$

We can then read out $\tilde{\mathbf{M}}$, and take the limit of $q \rightarrow 0$ for a uniform magnetic field.

Non-interacting periodic systems— For clarity, we first carry out the perturbation calculation for non-interacting periodic systems. The single-particle Hamiltonian can be expanded as $\hat{H} = \hat{H}_0 + \hat{V}_B$, where \hat{H}_0 is the unperturbed Hamiltonian, which yields the band dispersion $\epsilon_{n\mathbf{k}}$ and the corresponding Bloch wave function

$\psi_{n\mathbf{k}}(\mathbf{r}) = \exp(i\mathbf{k} \cdot \mathbf{r}) u_{n\mathbf{k}}(\mathbf{r})$, and \hat{V}_B denotes the coupling to the external magnetic field:

$$\hat{V}_B = \frac{e}{2} [\hat{\mathbf{v}} \cdot \mathbf{A}(\mathbf{r}) + \mathbf{A}(\mathbf{r}) \cdot \hat{\mathbf{v}}], \quad (6)$$

where $\hat{\mathbf{v}}$ is the velocity operator, and $\mathbf{A}(\mathbf{r})$ is the vector potential

$$\mathbf{A}(\mathbf{r}) = -B \frac{\sin qy}{q} \hat{x}, \quad (7)$$

which corresponds to the magnetic field discussed earlier. It is natural to define the grand-canonical ensemble energy density as $K(\mathbf{r}) = \sum_{n\mathbf{k}} f_{n\mathbf{k}} \text{Re}\{\psi_{n\mathbf{k}}^*(\mathbf{r}) \hat{K} \psi_{n\mathbf{k}}(\mathbf{r})\}$, where $\hat{K} = \hat{H} - \mu \hat{N}$, and $f_{n\mathbf{k}}$ are the occupation number of the single-electron states of band index n and crystal-momentum \mathbf{k} [19]. To first order in the perturbation, three kinds of terms arise from changes in the occupation number, the operator \hat{K} , and the wave function:

$$\delta K(\mathbf{r}) = \text{Re} \sum_{n\mathbf{k}} \left\{ \delta f_{n\mathbf{k}} \psi_{n\mathbf{k}}^* \hat{K}_0 \psi_{n\mathbf{k}} + f_{n\mathbf{k}} \psi_{n\mathbf{k}}^* \hat{V}_B \psi_{n\mathbf{k}} + f_{n\mathbf{k}} \left(\psi_{n\mathbf{k}}^* \hat{K}_0 \delta \psi_{n\mathbf{k}} + \delta \psi_{n\mathbf{k}}^* \hat{K}_0 \psi_{n\mathbf{k}} \right) \right\}. \quad (8)$$

The orbital magnetization can be determined from the appropriate Fourier component of the energy density, i.e.,

$$\tilde{M}_z = -\frac{2}{VB} \int d\mathbf{r} \delta K(\mathbf{r}) \cos qy. \quad (9)$$

It is easy to verify that the first two terms of Eq. (8) does not contribute to \tilde{M}_z , and only the contribution from the change of the wave functions remains. The first order perturbation to the wave function reads:

$$\delta \psi_{n\mathbf{k}}(\mathbf{r}) = -\frac{eB}{4iq} \sum_{n'} \left\{ \frac{e^{i(\mathbf{k}+\mathbf{q}) \cdot \mathbf{r}} |u_{n'\mathbf{k}+\mathbf{q}}\rangle \langle u_{n'\mathbf{k}+\mathbf{q}} | \hat{v}_x(\mathbf{k}) + \hat{v}_x(\mathbf{k}+\mathbf{q}) | u_{n\mathbf{k}}\rangle}{\epsilon_{n\mathbf{k}} - \epsilon_{n'\mathbf{k}+\mathbf{q}}} - (q \rightarrow -q) \right\}, \quad (10)$$

where $\hat{v}(\mathbf{k}) \equiv \partial \hat{H}_0(\mathbf{k}) / \partial (\hbar \mathbf{k})$ is the velocity operator, and $\hat{H}_0(\mathbf{k})$ is defined from the unperturbed Hamiltonian by shifting the momentum operator with $\hbar \mathbf{k}$. The transformed Hamiltonian acts on the periodic functions, with $u_{n\mathbf{k}}$ being its eigenfunctions and the band energy $\epsilon_{n\mathbf{k}}$ its eigenvalues. Making use Eqs. (8–10), we have:

$$\tilde{M}_z = \frac{e}{4q} \text{Im} \sum_{nn'\mathbf{k}} \frac{(\epsilon_{n\mathbf{k}} + \epsilon_{n'\mathbf{k}+\mathbf{q}} - 2\mu) \langle u_{n\mathbf{k}} | u_{n'\mathbf{k}+\mathbf{q}} \rangle \langle u_{n'\mathbf{k}+\mathbf{q}} | \hat{v}_x(\mathbf{k}) + \hat{v}_x(\mathbf{k}+\mathbf{q}) | u_{n\mathbf{k}} \rangle}{\epsilon_{n\mathbf{k}} - \epsilon_{n'\mathbf{k}+\mathbf{q}}} (f_{n\mathbf{k}} - f_{n'\mathbf{k}+\mathbf{q}}). \quad (11)$$

Taking the long-wavelength limit $q \rightarrow 0$, we obtain:

$$\begin{aligned} \tilde{M}_z = & \frac{e}{2} \text{Im} \sum_{n \neq n'\mathbf{k}} \frac{(\epsilon_{n\mathbf{k}} + \epsilon_{n'\mathbf{k}} - 2\mu) \langle u_{n\mathbf{k}} | \partial u_{n'\mathbf{k}} / \partial k_y \rangle \langle u_{n'\mathbf{k}} | \hat{v}_x(\mathbf{k}) | u_{n\mathbf{k}} \rangle}{\epsilon_{n\mathbf{k}} - \epsilon_{n'\mathbf{k}}} (f_{n\mathbf{k}} - f_{n'\mathbf{k}}) \\ & + e \sum_{n\mathbf{k}} (\epsilon_{n\mathbf{k}} - \mu) \text{Im} \left[v_x^n(\mathbf{k}) \left\langle u_{n\mathbf{k}} \left| \frac{\partial u_{n\mathbf{k}}}{\partial k_y} \right\rangle + \left\langle \frac{\partial u_{n\mathbf{k}}}{\partial k_y} \right| \hat{v}_x(\mathbf{k}) \right| u_{n\mathbf{k}} \right\rangle + \frac{1}{2} \left\langle u_{n\mathbf{k}} \left| \frac{\partial \hat{v}_x(\mathbf{k})}{\partial k_y} \right| u_{n\mathbf{k}} \right\rangle \right] f'_{n\mathbf{k}}, \end{aligned} \quad (12)$$

where $f'_{n\mathbf{k}} \equiv \partial f(\epsilon_{n\mathbf{k}}) / \partial \epsilon_{n\mathbf{k}}$. The second term comes from the intra-band contribution with $n = n'$. Eq. (12) can be further simplified with the help of the relations $\langle u_{n'\mathbf{k}} | \hat{v}_x(\mathbf{k}) | u_{n\mathbf{k}} \rangle = (1/\hbar)(\epsilon_{n\mathbf{k}} - \epsilon_{n'\mathbf{k}}) \langle u_{n'\mathbf{k}} | \partial u_{n\mathbf{k}} / \partial k_x \rangle$ for $n \neq n'$,

and $\text{Im}[\dots] = (1/\hbar)\text{Im}\langle\partial u_{n\mathbf{k}}/\partial k_y|\epsilon_n(\mathbf{k}) - \hat{H}_0(\mathbf{k})|\partial u_{n\mathbf{k}}/\partial k_x\rangle$, where $[\dots]$ denotes the expression inside the square bracket in Eq. (12). Combining these relations, and generalizing the result to the other components of $\tilde{\mathbf{M}}$, we obtain finally:

$$\tilde{\mathbf{M}} = -\frac{e}{2\hbar}i \sum_{n\mathbf{k}} \left\{ \left\langle \frac{\partial u_{n\mathbf{k}}}{\partial \mathbf{k}} \right| \left[\epsilon_n(\mathbf{k}) + \hat{H}_0(\mathbf{k}) - 2\mu \right] \times \left| \frac{\partial u_{n\mathbf{k}}}{\partial \mathbf{k}} \right\rangle f_{n\mathbf{k}} - (\epsilon_{n\mathbf{k}} - \mu) \left\langle \frac{\partial u_{n\mathbf{k}}}{\partial \mathbf{k}} \right| [\epsilon_n(\mathbf{k}) - \hat{H}_0(\mathbf{k})] \times \left| \frac{\partial u_{n\mathbf{k}}}{\partial \mathbf{k}} \right\rangle f'_{n\mathbf{k}} \right\}. \quad (13)$$

The auxiliary and proper orbital magnetization become the same at zero temperature. In this case, the second term in the above expression Eq. (13) vanishes because f' becomes a δ -function of $(\epsilon_{n\mathbf{k}} - \mu)$. The result is in perfect agreement with the semiclassical formula of zero temperature orbital magnetization of Xiao *et al.* [1]. For finite temperatures, we integrate Eq. (3) and obtain:

$$\mathbf{M} = \sum_{n\mathbf{k}} \left\{ \mathbf{m}_n(\mathbf{k}) f_{n\mathbf{k}} + \frac{e}{\hbar} \Omega_n(\mathbf{k}) \frac{1}{\beta} \ln \left(1 + e^{-\beta(\epsilon_{n\mathbf{k}} - \mu)} \right) \right\}, \quad (14)$$

where $\mathbf{m}_n(\mathbf{k}) \equiv (e/2\hbar)i\langle \nabla_{\mathbf{k}} u_{n\mathbf{k}} | [\epsilon_n(\mathbf{k}) - \hat{H}_0(\mathbf{k})] \times | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle$ is the orbital moment of state n, \mathbf{k} and $\Omega_n(\mathbf{k}) \equiv i\langle \nabla_{\mathbf{k}} u_{n\mathbf{k}} | \times | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle$ is the Berry curvature. The same expression was also obtained in Ref. [10].

Thus, all the previously known results are recovered by our fully quantum mechanical formalism. These results are valid not only for insulators with or without a Chern number, but also for metals at zero or finite temperatures. This implies that the semi-classical results are in general valid to linear order in the external fields. In hindsight, this should have been expected, because the semiclassical theory is designed to be exact in the limit of long length scales in the perturbation to the Hamiltonian. In our case, this length scale (through the vector potential) does diverge in the limit of vanishing magnetic field.

Generalization to interacting systems — It is very desirable to generalize the above results to an interacting system. This can be done exactly within the framework of the current and spin density functional theory (CSDFT) [6, 7]. CSDFT is a generalization of the spin density functional theory which includes the current density as an independent variable for the energy functional and thus provides direct access, via a variational principle of the Hohenberg-Kohn type, to the current density and the orbital magnetization of the thermodynamic equilibrium ensemble [11]. Following the formalism of Ref. [7], the many-body problem can be reduced to solving an effective one-body Schrödinger equation:

$$\left[\frac{1}{2m} (-i\hbar \nabla + e\mathbf{A}'_{\sigma}(\mathbf{r}))^2 + V'_{\sigma}(\mathbf{r}) \right] \psi_{i\sigma}(\mathbf{r}) = \epsilon_{i\sigma} \psi(\mathbf{r}), \quad (15)$$

with

$$V'_{\sigma} = V_{\sigma} + V_H + V_{xc\sigma} + \frac{e^2}{2m} (\mathbf{A}_{\sigma}^2 - \mathbf{A}'_{\sigma}{}^2), \quad (16)$$

$$\mathbf{A}'_{\sigma} = \mathbf{A}_{\sigma} + \mathbf{A}_{xc\sigma}. \quad (17)$$

Here V_{σ} and \mathbf{A}_{σ} are the external scalar and vector potential, respectively, acting on the σ component of the spin ($\sigma = \uparrow$ or \downarrow); $V_H = e^2 \int d\mathbf{r}' n(\mathbf{r}')/|\mathbf{r} - \mathbf{r}'|$ is the Hartree potential, and $V_{xc\sigma}$ and $\mathbf{A}_{xc\sigma}$ are the exchange-correlation (xc) scalar and vector potentials derived from the xc energy functional $\Omega_{xc}[n_{\sigma}, \mathbf{j}_{p\sigma}]$ according to the formulas $V_{xc\sigma} = \delta\Omega_{xc}/\delta n_{\sigma}$, $e\mathbf{A}_{xc\sigma} = \delta\Omega_{xc}/\delta \mathbf{j}_{p\sigma}$. The density, $n_{\sigma}(\mathbf{r})$, and the paramagnetic current density, $\mathbf{j}_{p\sigma}(\mathbf{r})$ are to be determined self-consistently from the solutions of the above equation according to the formulas $n_{\sigma}(\mathbf{r}) = \sum_i |\psi_{i\sigma}(\mathbf{r})|^2 f(\epsilon_{i\sigma})$ and $\mathbf{j}_{p\sigma}(\mathbf{r}) = (-i\hbar/2m) \sum_i [\psi_{i\sigma}^*(\mathbf{r}) \nabla \psi_{i\sigma}(\mathbf{r}) - \nabla \psi_{i\sigma}^*(\mathbf{r}) \psi_{i\sigma}(\mathbf{r})] f(\epsilon_{i\sigma})$. At finite temperature, the thermodynamic potential functional can be written as [7]:

$$\Omega = -\frac{1}{\beta} \sum_{i\sigma} \ln [1 + e^{-\beta(\epsilon_{i\sigma} - \mu)}] - \frac{1}{2} e^2 \iint d\mathbf{r} d\mathbf{r}' \frac{n(\mathbf{r})n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} - \sum_{\sigma} \int d\mathbf{r} n_{\sigma}(\mathbf{r}) V_{xc\sigma}(\mathbf{r}) - e \sum_{\sigma} \int d\mathbf{r} \mathbf{j}_{p\sigma}(\mathbf{r}) \cdot \mathbf{A}_{xc\sigma}(\mathbf{r}) + \Omega_{xc}[n_{\sigma}, \mathbf{j}_{p\sigma}], \quad (18)$$

which is a functional of four fields: the densities n_{σ} and $\mathbf{j}_{p\sigma}$, and the external potentials V_{σ} and \mathbf{A}_{σ} — the last two enter the expression through the eigenvalues $\epsilon_{i\sigma}$.

To calculate the orbital magnetization one needs to evaluate the variation $\delta\Omega$ of the thermodynamic potential in response to a variation of the external magnetic field, which, in turn, is generated by a variation in the external vector potentials $\delta\mathbf{A}_{\sigma}$. In general, $\delta\Omega$ can be separated into two contributions: the primary one ($\delta\Omega|_{n_{\sigma}, \mathbf{j}_{p\sigma}}$) arises directly from the variation of the vector potentials, keeping n_{σ} and $\mathbf{j}_{p\sigma}$ constant at their unperturbed values; the secondary one ($\delta\Omega|_{V_{\sigma}, \mathbf{A}_{\sigma}}$) might arise from the changes of n_{σ} and $\mathbf{j}_{p\sigma}$ at constant external potentials (these changes would affect Ω via the modification of the effective potentials V_H , $V_{xc\sigma}$ and $\mathbf{A}_{xc\sigma}$):

$$\delta\Omega = \delta\Omega|_{n_{\sigma}, \mathbf{j}_{p\sigma}} + \delta\Omega|_{V_{\sigma}, \mathbf{A}_{\sigma}}. \quad (19)$$

It is easy to see that $\delta\Omega|_{n_{\sigma}, \mathbf{j}_{p\sigma}}$ contributes an orbital magnetization that is exactly given by Eq. (14), as if the system were a noninteracting system with eigenfunctions and eigenvalues determined by Eq. (15). This is because the variation of the external vector potential affects only the eigenvalues $\epsilon_{i\sigma}$ in the one-body term of Eq. (18). To evaluate $\delta\Omega|_{V_{\sigma}, \mathbf{A}_{\sigma}}$ it is sufficient to observe that for given external potentials the thermodynamic potential Ω

is stationary against small changes of the density and the current about their equilibrium values: $\delta\Omega/\delta n_\sigma = \delta\Omega/\delta \mathbf{j}_{p\sigma} = 0$.

Thus we have

$$\delta\Omega|_{V_\sigma, \mathbf{A}_\sigma} = 0. \quad (20)$$

We then conclude that in the context of the CSDFT, we can treat the system as an effective one-body system, and use Eq. (14) to calculate the orbital magnetization, albeit using the dispersion and wave-functions derived from Eq. (15).

We stress that this conclusion would not hold true if the one-electron orbitals and their energies were calculated within the framework of the *ordinary* density functional theory [15] (as opposed to CSDFT). In such a formulation the xc energy functional would be a functional of density *and* magnetic field: $\Omega_{xc}[n_\sigma, \mathbf{B}]$. Then the formula for the orbital magnetization would include a functional derivative of Ω_{xc} with respect to \mathbf{B} , at variance to the one-electron formula of Eq. (14).

Finite fields — Our formula can be applied rigorously to finite magnetic fields if these fields are rational in the sense that fluxes through the faces of a unit cell are fractional multiples of the flux quantum h/e . In this case, one can define Bloch like eigen-states with respect to magnetic translations, which are ordinary translations on the crystal combined with gauge transformations. Orbital magnetization at such a field can then be calculated perturbatively by adding a small change δB to this field. Both the semiclassical theory and quantum perturbation with respect to δB give the same expression for the orbital magnetization, provided we use the magnetic Bloch wavefunctions as a basis. Like wise, the CSDFT can also be formulated for systems with periodic boundary condi-

tions with respect to magnetic translations, so the justification of our results for interacting systems is straightforward [7]. Indeed, current density functional theory has been used to study the formation of an electron crystal (Wigner crystal) at very high magnetic field [16].

The situation for irrational fields is a bit tricky if one insists on rigorous results [17]. One may consider an irrational field as a limit of sequence of rational fields. This is possible if the magnetization depends on the field continuously, which is expected to be the case when the temperature is finite. Indeed, when the fast de Haas-van Alphen oscillations are smeared out, the average magnetization changes continuously with the Fermi energy [17]. For a fixed Fermi energy, the average magnetization also changes continuously with the magnetic field. Therefore, we expect that the average magnetization is a continuous function of magnetic field at a fixed density of electrons.

In summary, we have presented a full quantum derivation of the orbital magnetization formula. The derivation is generally valid for insulators with or without a Chern number, for metals at zero or finite temperatures, and at weak as well as strong magnetic fields. We also find that the resulting formula is directly applicable to interacting systems provided one uses one-electron energies and wave functions obtained from the self-consistent solution of the Kohn-Sham equation of current and spin-density functional theory.

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- [1] D. Xiao, J. Shi, and Q. Niu, Phys. Rev. Lett. **95**, 137204 (2005).
 - [2] M.-C. Chang and Q. Niu, Phys. Rev. Lett. **75**, 1348 (1995).
 - [3] G. Sundaram and Q. Niu, Phys. Rev. B **59**, 14915 (1999).
 - [4] T. Thonhauser, D. Ceresoli, D. Vanderbilt, and R. Resta, Phys. Rev. Lett. **95**, 137205 (2005).
 - [5] D. Ceresoli, T. Thonhauser, D. Vanderbilt, and R. Resta, Phys. Rev. B **74**, 024408 (2006).
 - [6] G. Vignale and M. Rasolt, Phys. Rev. Lett. **59**, 2360 (1987).
 - [7] G. Vignale and M. Rasolt, Phys. Rev. B **37**, 10685 (1988).
 - [8] G. Vignale, Phys. Rev. Lett. **67**, 358 (1991).
 - [9] F. Mauri and S. G. Louie, Phys. Rev. Lett. **76**, 4246 (1996).
 - [10] D. Xiao, Y. Yao, Z. Fang, and Q. Niu, Phys. Rev. Lett. **97**, 026603 (2006).
 - [11] H. Ebert, M. Battocletti, and E. K. U. Gross, Europhys. Lett. **40**, 545 (1997).
 - [12] M. Richter, Handbook of magnetic materials, edited by H. H. J. Buschow (Elsevier) **13**, 87 (2001).
 - [13] S. Rohra and A. Görling, Phys. Rev. Lett. **97**, 013005 (2006).
 - [14] S. Pittalis, S. Kurth, N. Helbig, and E. K. U. Gross, Phys. Rev. A **74**, 062511 (2006).
 - [15] C. J. Grayce and R. A. Harris, Phys. Rev. A **50**, 3089 (1994).
 - [16] G. Vignale, Phys. Rev. B **47**, 10105 (1993).
 - [17] O. Gat and J. E. Avron, Phys. Rev. Lett. **91**, 186801 (2003).
 - [18] The separation of spin and orbital degrees of freedom is only possible in a semi-relativistic framework, such as the one provided by the Pauli equation. In a fully relativistic treatment orbital and spin magnetizations merge together, and our formula gives the total magnetization.
 - [19] The definition here differs slightly in the kinetic energy density from the conventional one by a total divergence. Its perturbation by the magnetic field vanishes in the limit of $q \rightarrow 0$, and therefore does not affect our final result.